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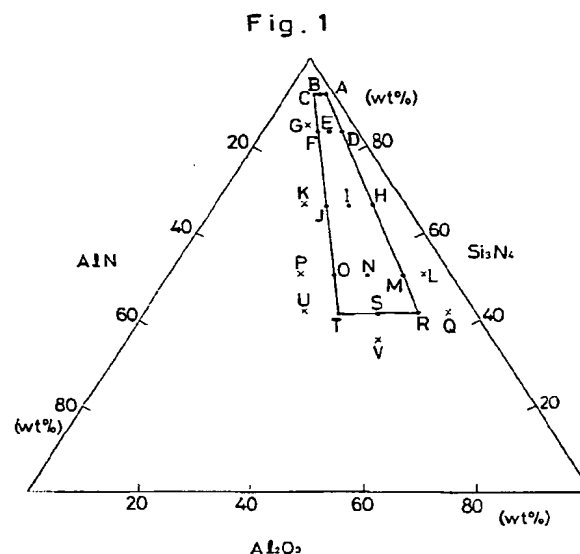
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**A sintered ceramic body for a spark plug insulator and method of sintering the same.**

In a sintered ceramic body used for a spark plug insulator, the ceramic body has silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride ( $\text{AlN}$ ) and alumina ( $\text{Al}_2\text{O}_3$ ). Reduced weight percentage of the silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride ( $\text{AlN}$ ) and alumina ( $\text{Al}_2\text{O}_3$ ) in the sintered ceramic body falls within an area surrounded by points A (91.5, 1.5, 7.0), B (91.5, 2.5, 6.0), C (91.5, 3.5, 5.0), F (83.0, 7.0, 10.0), J (66.0, 14.0, 20.0), O (50.0, 20.5, 29.0), T (41.0, 24.0, 35.0), S (41.0, 17.0, 42.0), R (41.0, 10.0, 49.0), M (50.0, 8.5, 41.5), H (66.0, 6.0, 28.0), D (83.0, 3.0, 14.0) and A (91.5, 1.5, 7.0) at a three-component diagram. As one sintering additive, yttrium oxide ( $\text{Y}_2\text{O}_3$ ) is selected, weight percentage of the sintering additive ranges from 0.3 % to 15.0 % of the total weight of the silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride ( $\text{AlN}$ ) and alumina ( $\text{Al}_2\text{O}_3$ ).



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This invention relates to a sintered ceramic body well-suited for a spark plug insulator which needs an elevated insulation property at high ambient temperature with good thermal conductivity.

In a spark plug insulator for an internal combustion engine, a oxinite sintered ceramic body has been employed since the sintered ceramic body has good thermal conductivity.

The oxinite sintered ceramic body, however, comes to decrease its electrical insulation when exposed to high ambient temperature, and grows crystals treeing over the surface of the sintered ceramic body when high voltage is applied.

Therefore, it is an aim of the invention to provide a sintered ceramic body and spark plug insulator which are capable of maintaining an elevated insulation property at high ambient temperature with good thermal conductivity.

According to the invention, there is provided a sintered ceramic body for a spark plug insulator comprising silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride ( $\text{AlN}$ ) and alumina ( $\text{Al}_2\text{O}_3$ ), reduced weight percentage of the silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride ( $\text{AlN}$ ) and alumina ( $\text{Al}_2\text{O}_3$ ) in the sintered ceramic body falling within an area surrounded by points A (91.5, 1.5, 7.0), B (9.15, 2.5, 6.0), C (91.5, 3.5, 5.0), F (83.0, 7.0, 10.0), J (66.0, 14.0, 20.0), O (50.0, 20.5, 29.0), T (41.0, 24.0, 35.0), S (41.0, 17.0, 42.0), R (41.0, 10.0, 49.0), M (50.0, 8.5, 41.5), H (66.0, 6.0, 28.0), D (83.0, 3.0, 14.0) and A (91.5, 1.5, 7.0) at a three-component diagram in which numerals in parentheses indicate the reduced weight percentage of the silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride ( $\text{AlN}$ ) and alumina ( $\text{Al}_2\text{O}_3$ ) respectively; and at least one sintering additive selected from the group consisting yttrium oxide ( $\text{Y}_2\text{O}_3$ ), calcium oxide ( $\text{CaO}$ ), lanthanum oxide ( $\text{La}_2\text{O}_3$ ), magnesia ( $\text{MgO}$ ), cerium oxide ( $\text{CeO}_2$ ), scandium oxide ( $\text{Sc}_2\text{O}_3$ ), strontium oxide ( $\text{SrO}$ ), barium oxide ( $\text{BaO}$ ) and neodymium ( $\text{Nd}_2\text{O}_3$ ), weight percentage of the sintering additive ranging from 0.3 % to 15.0 % of the total weight of the silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride ( $\text{AlN}$ ) and alumina ( $\text{Al}_2\text{O}_3$ ).

Since an oxinite sintered ceramic is chemically structured by a strong covalent bondage, free electrons cause to represent electrical conductivity. An increased ambient temperature facilitates the movement of the free electrons to decrease an electrical resistance so as to reduce an insulation property of the sintered body.

The reduced weight percentage of the silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride ( $\text{AlN}$ ) and alumina ( $\text{Al}_2\text{O}_3$ ) is such that the ceramic sintered body is in the form of  $\beta$ -sialon ( $\text{Si}_m\text{Al}_n\text{N}_x\text{O}_y$ ), a substitutional solid solution in which (Si) of  $\text{Si}_3\text{N}_4$  is partly replaced by (Al), while (N) of  $\text{Si}_3\text{N}_4$  partly replaced by (O). The  $\beta$ -sialon is characterized by ion bondage, and has a chemical property similar to alumina ( $\text{Al}_2\text{O}_3$ ) which substantially maintains an uniform electrical resistance under high temperature.

The sintering additive of less than 0.3 weight percent brings an adverse affect on the sintering to decrease a density of the sintered body, and imparting a liquid-absorbing property. The sintering additive exceeding 15 weight percent forms vitrified phase on a granular boundary of the sintered body so as to significantly deteriorate its strength.

With the oxinite ceramic body employed to the spark plug insulator, there is provided a spark plug which is capable of imparting anti-fouling and heat-resistant property by maintaining an elevated insulation property and good thermal conductivity at high ambient temperature.

The present invention will be further described hereinafter with reference to the following description of exemplary embodiments and the accompanying drawings, in which:-

Fig. 1 is a three-component diagram of silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride ( $\text{AlN}$ ) and alumina ( $\text{Al}_2\text{O}_3$ ) depicted to show each weight-percentage area which keeps an electrical resistance of a sintered ceramic body 50 M $\Omega$  or more at 700 °C; and

Fig. 2 is a schematic plan view showing a device to measure insulation resistance of test pieces at high temperature.

Referring to Figs. 1, 2 and Table 1, an experiment is carried out in order to seek a weight-percentage area of a sintered body including silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride ( $\text{AlN}$ ) and alumina ( $\text{Al}_2\text{O}_3$ ) which keeps its electrical resistance 50 M $\Omega$  or more at 700 °C. The reference notations (A) ~ (V) in Fig. 1 corresponds to those of test pieces in Table 1.

(1) Silicon nitride ( $\text{Si}_3\text{N}_4$ ) powder is prepared whose granular size measures 1.5  $\mu\text{m}$  in average diameter (sedimentation analysis) with an oxygen-laden rate as 1.0 weight percent. Aluminum nitride ( $\text{AlN}$ ), alumina ( $\text{Al}_2\text{O}_3$ ) and yttrium oxide ( $\text{Y}_2\text{O}_3$ ) powder are added to the silicon nitride ( $\text{Si}_3\text{N}_4$ ) powder to form a mixture. Size of the aluminum nitride powder measures 1.0  $\mu\text{m}$  in average diameter (sedimentation analysis) with an oxygen-laden rate as 10 weight percent, while size of the alumina powder measures 1.0  $\mu\text{m}$  in average diameter. Test pieces hereinafter obtained are prepared by previously adopting each amount of ( $\text{Si}_3\text{N}_4$ ), ( $\text{Al}_2\text{O}_3$ ) and ( $\text{AlN}$ ) substantially according to Table 1 depicted hereinafter.

Each of sintering additives employed herein is 99.9 % purity with ratio of surface area to weight as 12 m<sup>2</sup>/g. The sintering additive is selected alone or combination from the group consisting of yttrium oxide ( $\text{Y}_2\text{O}_3$ ), calcium oxide ( $\text{CaO}$ ), lanthanum oxide ( $\text{La}_2\text{O}_3$ ), magnesia ( $\text{MgO}$ ), cerium oxide ( $\text{CeO}_2$ ), scandium oxide

( $\text{Sc}_2\text{O}_3$ ), strontium oxide ( $\text{SrO}$ ), barium oxide ( $\text{BaO}$ ) and neodymium ( $\text{Nd}_2\text{O}_3$ ).

An ethanol or wax-related binder is added to the mixture of the aluminum nitride, alumina, silicon nitride and yttrium oxide powder, and are kneaded within a nylon pot by means of a ball for approximately 24 hours.

A quantity of the silicon nitride ( $\text{Si}_3\text{N}_4$ ) and yttrium oxide ( $\text{Y}_2\text{O}_3$ ) powder is determined by taking the fact into consideration that 20 ~ 30 % of (Si) and (Y) disappear during a sintering process described hereinafter.

(2) After spray drying a slurry produced by kneading the mixture the mixture is formed into a compact plate which measures 50 mm in diameter and 3.0 mm in thickness. Then the compact plate is primarily sintered at about 500 °C, and pressed under 2 tons/cm<sup>2</sup> pressure by means of cold isostatic press (CIP).

(3) The compact plate is placed in a shell which is made of boron nitride (BN), and is secondarily sintered for 2 ~ 5 hours at temperature of 1650 °C ~ 1750 °C, while circulating nitrogen gas circulating within the shell according to sintering conditions shown in Table 1.

(4) The compact plate thus secondarily sintered (sintered body) is milled to measure 40 mm in diameter and 1.0 mm in thickness to provide test pieces (A) ~ (V). Each weight percentage of the silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride (AlN), alumina ( $\text{Al}_2\text{O}_3$ ) and yttrium oxide ( $\text{Y}_2\text{O}_3$ ) is measured on the basis of fluorescent-sensitive X-ray detection.

In this instance, each weight percentage of the silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride (AlN) and alumina ( $\text{Al}_2\text{O}_3$ ) is reduced from the sintered body as is referred to as reduced weight percentage hereinafter.

Each insulation resistance of ( $\text{Si}_3\text{N}_4$ ), (AlN), ( $\text{Al}_2\text{O}_3$ ) and ( $\text{Y}_2\text{O}_3$ ) is measured at 700 °C by using a device shown in Fig. 2 which has brass-made electrodes 100, 200, a heater 300 and a 500-volt digital resistance meter 400.

As a result of the experiment (4), it is found from Table 1 that test pieces (A) ~ (F), (H) ~ (J), (M) ~ (O) and (R) ~ (T) represent their insulation resistance of 50 MΩ or more at 700 °C.

An experiment is carried out to determine reduced weight-percentage area of the sintering additive which enables densely sintered body so as to sufficiently resist against fracture.

Table 1

test piece	reduced weight percentage of $\text{Si}_3\text{N}_4$ (wt%)	reduced weight percentage of $\text{AlN}$ (wt%)	reduced weight percentage of $\text{Al}_2\text{O}_3$ (wt%)	sintering conditions ( $^{\circ}\text{C} \times \text{Hr}$ )	insulation resistance at $700^{\circ}\text{C}$ (M $\Omega$ )
A	91.5	1.5	7.0	1750X2	85
B	91.5	2.5	6.0	1750X2	100
C	91.5	3.5	5.0	1750X2	90
D	83.0	3.0	14.0	1700X2	110
E	83.0	5.0	12.0	1700X2	120
F	83.0	7.0	10.0	1700X5	170
G	83.0	9.0	8.0	1700X5	30*
H	66.0	6.0	28.0	1700X2	150
I	66.0	10.0	24.0	1700X2	180
J	66.0	14.0	20.0	1700X2	200
K	66.0	18.0	16.0	1700X2	20*
L	50.0	5.0	45.0	1700X2	15*
M	50.0	8.5	41.5	1650X2	120
N	50.0	14.5	35.5	1650X2	120
O	50.0	20.5	29.0	1650X2	65
P	50.0	26.5	22.5	1650X2	20*
Q	41.0	5.0	54.0	1650X2	25*
R	41.0	10.0	49.0	1650X2	500
S	41.0	17.0	42.0	1650X2	320
T	41.0	24.0	35.0	1650X2	190
U	41.0	31.0	28.0	1700X2	30*
V	35.0	20.0	45.0	1650X2	45*

Auxiliary agent used at above test pieces is  $\text{Y}_2\text{O}_3$  weight percentage of which is 5 wt% of the total weight of  $\text{Si}_3\text{N}_4$ ,  $\text{AlN}$  and  $\text{Al}_2\text{O}_3$ .  
 test pieces A~F, H~J, M~O, R~T: according to the invention.  
 Counterpart test pieces: G, K, L, P, Q, U, V (vulnerable to treeing due to low insulation resistance at  $700^{\circ}\text{C}$  as indicated by asterisk \*).

The slurry spray dried in the same manner as item (1) is formed into two shape of a disk and plate. The disk measures 50 mm in diameter and 3.0 mm in thickness, while the plate measures 10 mm x 40 mm, and 5.0 mm in thickness. The disk and plate are primarily sintered in the same manner as item (1), and placed in a shell which is made of boron nitride (BN) employed for its high refractory. Then the disk and plate are secondarily sintered for 2 ~ 5 hours at temperature of 1550 °C ~ 1750 °C, while circulating nitrogen gas circulating within the shell according to sintering conditions shown in Table 2. The disk is milled to measure 40 mm in diameter and 1.0 mm in thickness, while the plate is milled to measure 8 mm x 30 mm and 4.0 mm in thickness to provide test pieces.

By using these test pieces, reduced weight percentage of the sintering additive is measured on the basis of fluorescent-sensitive X-ray detection, while each relative density of the test pieces is calculated by comparing theoretical density with apparent density measured by Archimedes' method. Three-point bending test according to JIS C4104 is used to measure resistant strenght of the test pieces against fracture in addition to each reduced weight percentage of (Si<sub>3</sub>N<sub>4</sub>), (AlN), (Al<sub>2</sub>O<sub>3</sub>) and insulation resistance of the test pieces.

As a result, it is found from Table 2 that the test pieces (a) ~ (h) have high relative density, high resistant strength against fracture and insulation resistance of 50 MΩ or more reduced weight percentage of these test pieces (a) ~ (h) ranges from 0.3 wt % to 15 wt % of the total weight of (Si<sub>3</sub>N<sub>4</sub>), (AlN) and (Al<sub>2</sub>O<sub>3</sub>).

Table 2

test piece	reduced weight percentage of SiN <sub>4</sub> (wt%)	reduced weight percentage of AlN (wt%)	reduced weight percentage of Al <sub>2</sub> O <sub>3</sub> (wt%)	weight percentage of sintering additive (wt%)	sintering conditions (T x Hr)	relative density (%)	resistant strength against fracture (kg/mm <sup>2</sup> )
a	91.5	2.5	6.0	Y <sub>2</sub> O <sub>3</sub> 10	1650x2	99	52
b	83.0	7.0	10.0	Y <sub>2</sub> O <sub>3</sub> 15	1650x2	99	45
c	83.0	7.0	10.0	CaO 3	1650x2	99	42
d	50.0	14.5	35.5	MgO 2	1600x2	98	24
e	41.0	17.0	42.0	CeO <sub>2</sub> 7	1700x2	99	22
f	41.0	16.5	9.5	Y <sub>2</sub> O <sub>3</sub> 5	1650x2	99	26
g	83.0	3.0	7.0	Y <sub>2</sub> O <sub>3</sub> 0.3	1750x2	99	32
h	90.0	5.0	12.0	CeO <sub>2</sub> 0.5	1700x2	99	51
i	83.0	5.0	12.0	CeO <sub>2</sub> 0.1	1750x2	92*	not measured
j	66.0	6.0	28.0	CeO <sub>2</sub> 0.2	1750x2	93*	not measured
k	66.0	6.0	28.0	CeO <sub>2</sub> 0.15	1750x2	91*	not measured
l	66.0	6.0	28.0	CeO <sub>2</sub> 18	1550x2	99	16**
m	66.0	6.0	28.0	CeO <sub>2</sub> 21	1600x2	99	18**

Insulation resistance: 50MΩ or more for all the test pieces

test pieces (a)~(h): according to the invention

test pieces (i)~(m): counterpart specimens

(\*) : representing liquid-absorbing property due to low relative density and low resistant strength against fracture.

(\*\*) : representing very low resistant strength against fracture.

These the test piece (a) ~ (h) have good insulation resistance and favorable thermal conductivity at the temperature of 700 °C, thus making the sintered body well-suited for a spark plug insulator since the spark plug insulator needs the thermal conductivity of more than 90 W/m and the insulation resistance of more than 50 MΩ at 700 °C from treeing-preventing viewpoint.

5 Reduced weight percentage of ( $\text{Si}_3\text{N}_4$ ), (AlN) and ( $\text{Al}_2\text{O}_3$ ) of these test pieces (a) ~ (h) falls within an area surrounded by points A (91.5, 1.5, 7.0), B (91.5, 2.5, 6.0), C (91.5, 3.5, 5.0), F (83.0, 7.0, 10.0), J (66.0, 14.0, 20.0), O (50.0, 20.5, 29.0), T (41.0, 24.0, 35.0), S (41.0, 17.0, 42.0), R (41.0, 10.0, 49.0), M (50.0, 8.5, 41.5), H (66.0, 6.0, 28.0), D (83.0, 3.0, 14.0) and A (91.5, 1.5, 7.0) at three-component diagram shown in Fig. 1. Numerals in the parentheses indicate reduced weight percentage of silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride (AlN) and alumina ( $\text{Al}_2\text{O}_3$ ) in turn.

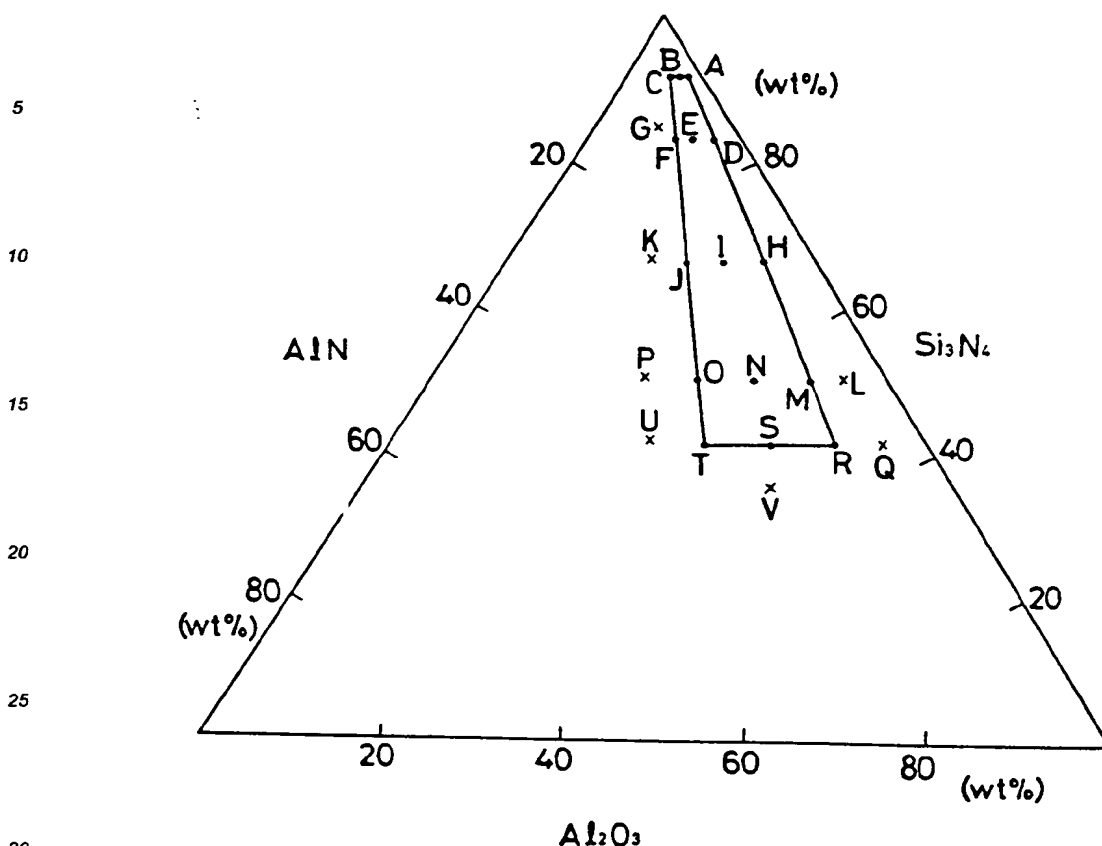
10 Therefore, it is necessary to arrange that the reduced weight percentage of the silicon nitride ranging from 41.0 % to 91.5 %, while reduced weight percentage of the aluminum nitride ranging from 2.5 % to 24.0 %, and the reduced weight percentage of the alumina ranging from 5.1 % to 49.0 % so as to maintain an electrical resistance of a sintered ceramic body 50 MΩ or more at 700 °C.

15 A spark plug insulator is made in accordance with the test pieces (a) ~ (h). After a center electrode, a resistor and a terminal electrode are placed within an axial bore of the insulator through an electrically conductive glass sealant, the insulator is placed within a metallic shell to form a spark plug which is found to ensure anti-fouling and heat-resistant property so as to prevent misfire.

20 While the invention has been described with reference to the specific embodiments, it is understood that this description is not to be construed in a limiting sense in as much as various modifications and additions to the specific embodiments may be made by skilled artisan without departing from the spirit and scope of the invention.

## 25 Claims

1. A spark plug insulator having a sintered ceramic body comprising silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride (AlN) and alumina ( $\text{Al}_2\text{O}_3$ ), reduced weight percentage of the silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride (AlN) and alumina ( $\text{Al}_2\text{O}_3$ ) in the sintered ceramic body falling within an area surrounded by points A (91.5, 1.5, 7.0), B (91.5, 2.5, 6.0), C (91.5, 3.5, 5.0), F (83.0, 7.0, 10.0), J (66.0, 14.0, 20.0), O (50.0, 20.5, 29.0), T (41.0, 24.0, 35.0), S (41.0, 17.0, 42.0), R (41.0, 10.0, 49.0), M (50.0, 8.5, 41.5), H (66.0, 6.0, 28.0), D (83.0, 3.0, 14.0) and A (91.5, 1.5, 7.0) at a three-component diagram listed below in which numerals in parentheses indicate the reduced weight percentage of the silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride (AlN) and alumina ( $\text{Al}_2\text{O}_3$ ) respectively; and



at least one wintering additive selected from the group consisting yttrium oxide ( $\text{Y}_2\text{O}_3$ ), calcium oxide ( $\text{CaO}$ ), lanthanum oxide ( $\text{La}_2\text{O}_3$ ), magnesia ( $\text{MgO}$ ), cerium oxide ( $\text{CeO}_2$ ), scandium oxide ( $\text{Sc}_2\text{O}_3$ ), strontium oxide ( $\text{SrO}$ ), barium oxide ( $\text{BaO}$ ) and neodymium ( $\text{Nd}_2\text{O}_3$ ), weight percentage of the sintering additive ranging from 0.3 % to 15.0 % of the total weight of the silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride ( $\text{AlN}$ ) and alumina ( $\text{Al}_2\text{O}_3$ ).

2. A method of sintering a ceramic body for a spark plug insulator comprising steps of:

preparing a mixture of silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride ( $\text{AlN}$ ) and alumina ( $\text{Al}_2\text{O}_3$ ), reduced weight percentage of the silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride ( $\text{AlN}$ ) and alumina ( $\text{Al}_2\text{O}_3$ ) in the sintered ceramic body falling within an area surrounded by points A (91.5, 1.5, 7.0), B (91.5, 2.5, 6.0), C (91.5, 3.5, 5.0), F (83.0, 7.0, 10.0), J (66.0, 14.0, 20.0), O (50.0, 20.5, 29.0), T (41.0, 24.0, 35.0), S (41.0, 17.0, 42.0), R (41.0, 10.0, 49.0), M (50.0, 8.5, 41.5), H (66.0, 6.0, 28.0), D (83.0, 3.0, 14.0) and A (91.5, 1.5, 7.0) at a three-component diagram listed below in which numerals in parentheses indicate the reduced weight percentage of the silicon nitride ( $\text{Si}_3\text{N}_4$ ), aluminum nitride ( $\text{AlN}$ ) and alumina ( $\text{Al}_2\text{O}_3$ ) respectively;

kneading the mixture for approximately 24 hours with addition of yttrium oxide ( $\text{Y}_2\text{O}_3$ ) and ethanol-based binder;

primarily sintering the kneaded mixture for approximately 2 hours at temperature of about 500 °C, and pressing the mixture under pressure of 2 tons/cm<sup>2</sup> to form a compact body;

placing the compact body in a shell which is made of boron nitride (BN), and secondarily sintering the compact body for 2 ~ 5 hours at temperature of 1650 °C ~ 1750 °C, while circulating nitrogen gas within the shell, so that reduced weight percentage of the silicon nitride in the sintered ceramic body ranging from 41.0 % to 91.5% inclusive, while reduced weight percentage of the aluminum nitride in the sintered ceramic body ranging from 2.5 % to 24.0 % inclusive, reduced weight percentage of the alumina in the sintered ceramic body ranging from 5.7 % to 49.0 % inclusive.



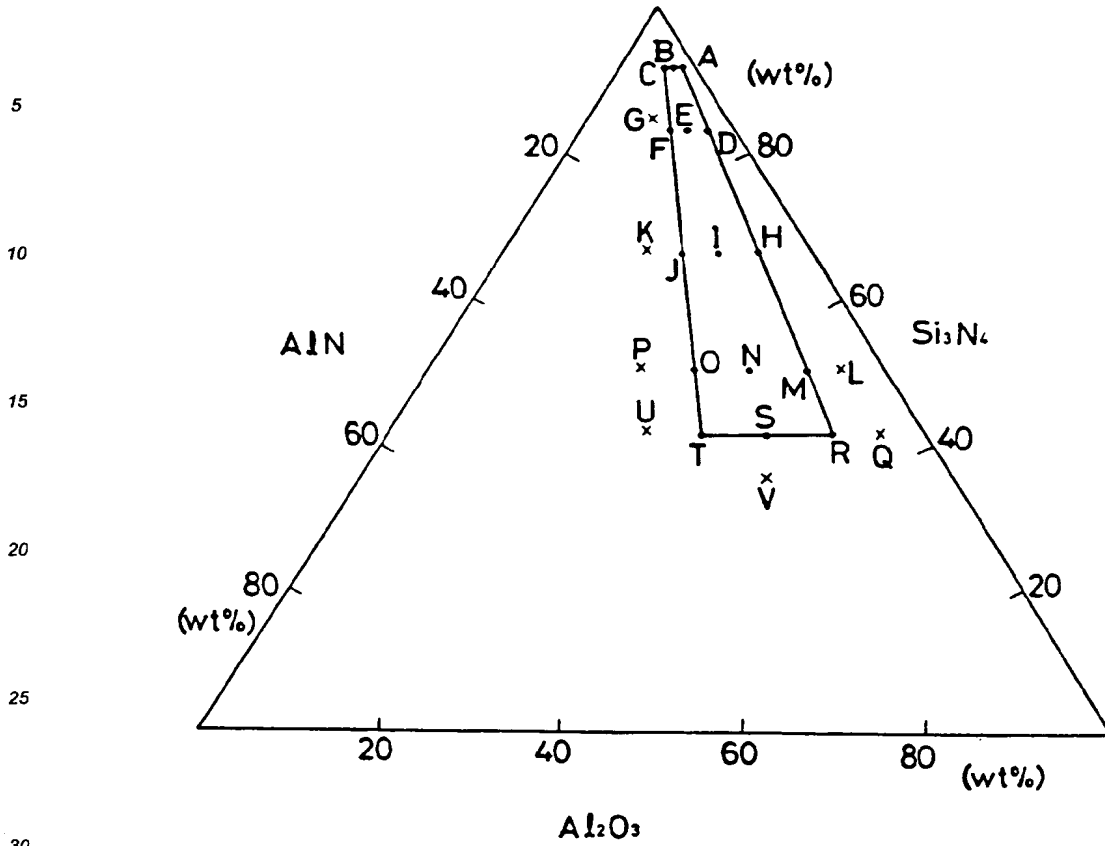


Fig. 1

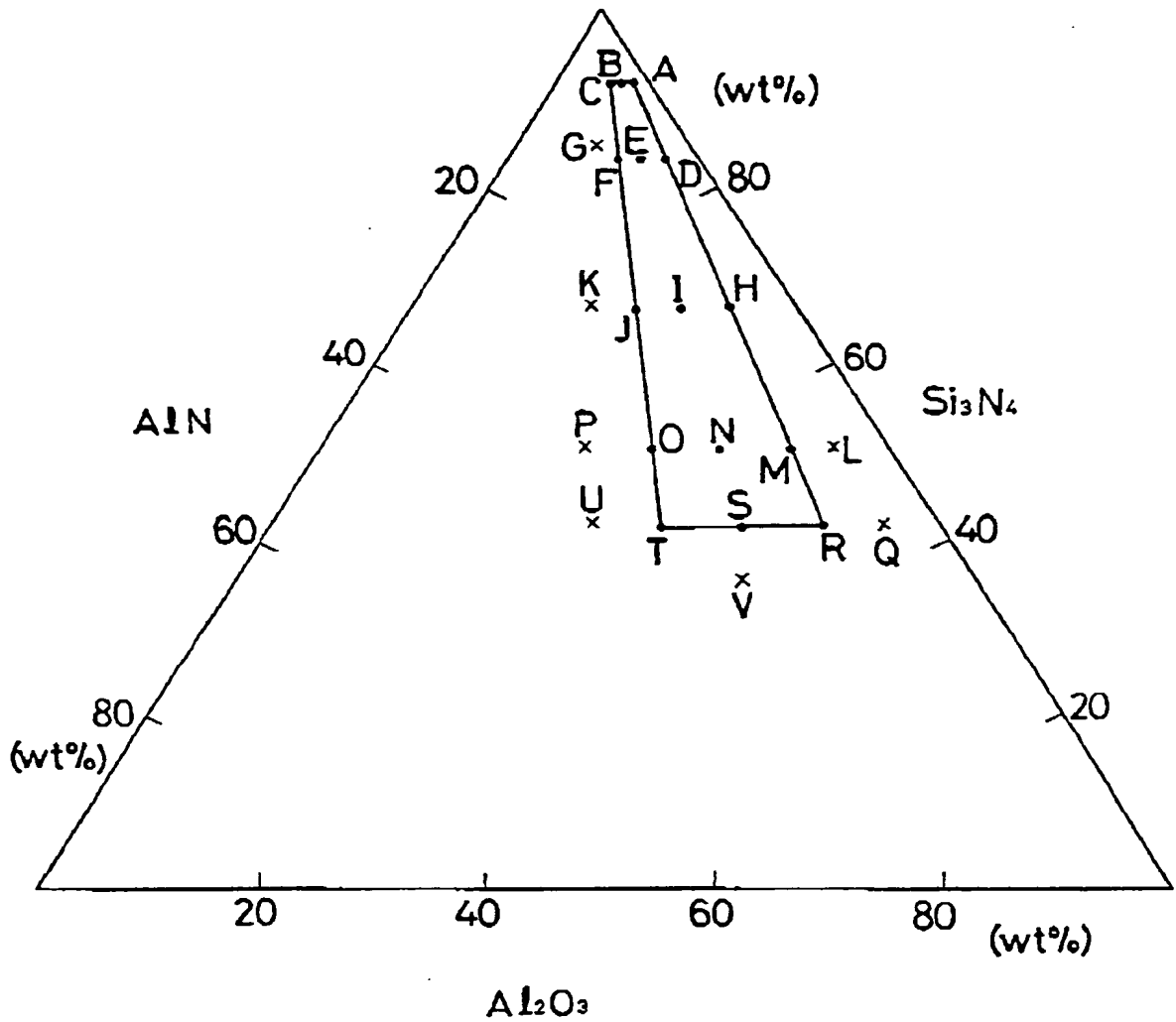
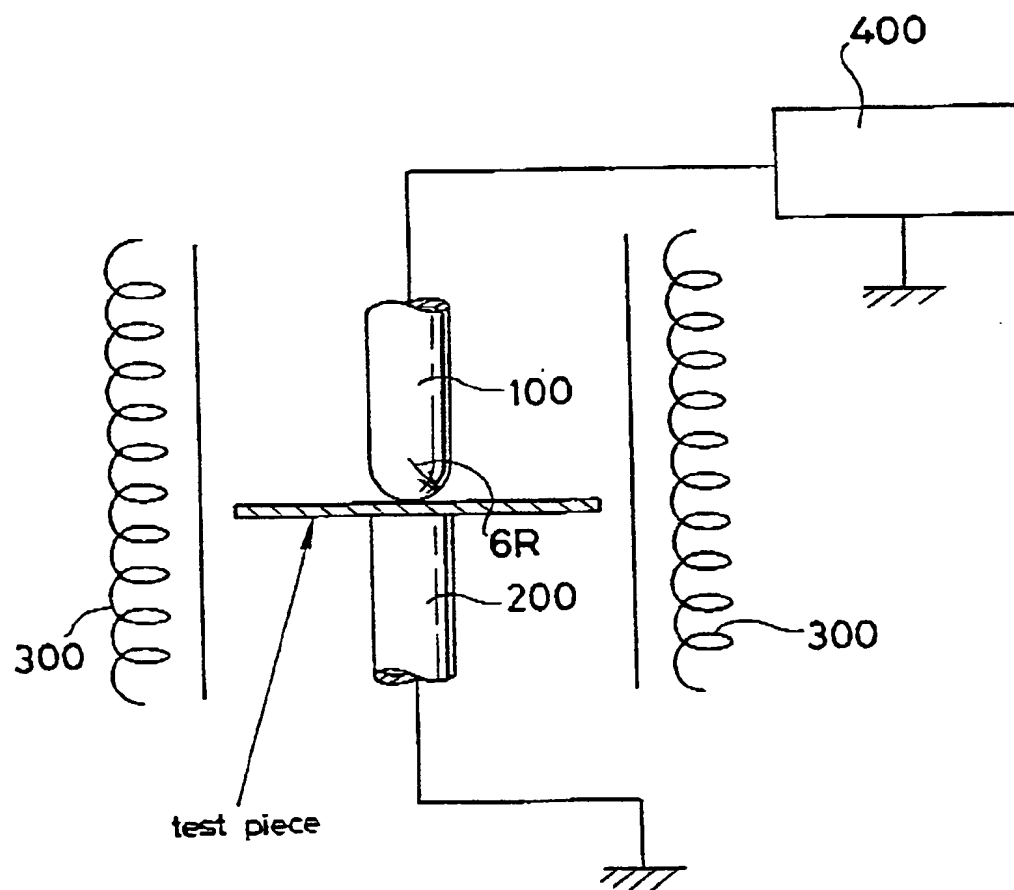


Fig. 2





European Patent  
Office

# EUROPEAN SEARCH REPORT

Application Number

EP 91 30 9769

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
X	EP-A-0 015 421 (TOKYO SHIBAURA DENKI KABUSHIKI KAISHA) * page 2, line 6 - line 11; claims 1-9; examples 19, 39 *	1, 2	C04B35/58 H01T13/38
A	US-A-4 845 061 (K. INOBUCHI ET AL.) * claims 2-4 *	1, 2	
			TECHNICAL FIELDS SEARCHED (Int. Cl.5)
			C04B H01T
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 29 JANUARY 1992	Examiner HAUCK H. N.
<p><b>CATEGORY OF CITED DOCUMENTS</b></p> <p>X : particularly relevant if taken alone  Y : particularly relevant if combined with another document of the same category  A : technological background  O : non-written disclosure  P : intermediate document</p> <p>T : theory or principle underlying the invention  E : earlier patent document, but published on, or after the filing date  D : document cited in the application  L : document cited for other reasons  &amp; : member of the same patent family, corresponding document</p>			

EPO FORM 150 (01/82) (P0401)